

(19)



Europäisches Patentamt  
European Patent Office  
Office européen des brevets



(11)

**EP 0 866 110 A1**

(12)

**EUROPEAN PATENT APPLICATION**

(43) Date of publication:  
23.09.1998 Bulletin 1998/39

(51) Int Cl.<sup>6</sup>: C09K 11/06, H05B 33/14,  
G03G 5/06, C07C 211/54

(21) Application number: 98301986.0

(22) Date of filing: 17.03.1998

(84) Designated Contracting States:  
AT BE CH DE DK ES FI FR GB GR IE IT LI LU MC  
NL PT SE  
Designated Extension States:  
AL LT LV MK RO SI

• Tamano, Michiko  
3-13, Kyobashi 2-chome, Chuo-ku, Tokyo (JP)  
• Okutsu, Satoshi  
3-13, Kyobashi 2-chome, Chuo-ku, Tokyo (JP)  
• Enokida, Toshio  
3-13, Kyobashi 2-chome, Chuo-ku, Tokyo (JP)

(30) Priority: 17.03.1997 JP 62568/97

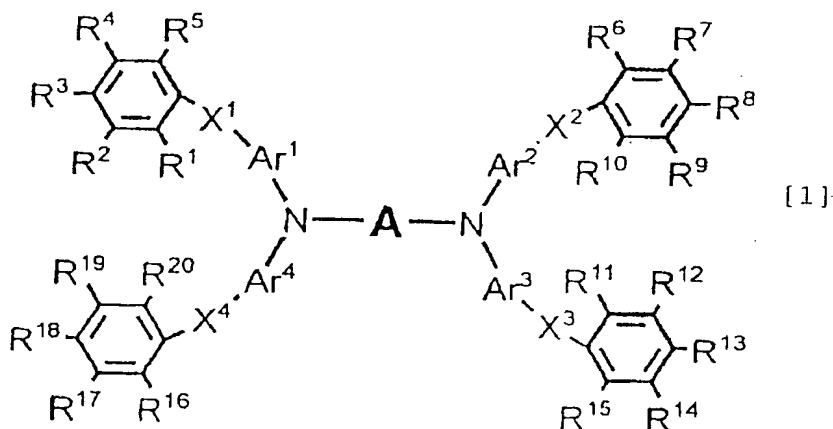
(71) Applicant: TOYO INK MANUFACTURING CO.,  
LTD.  
Tokyo (JP)

(74) Representative: Woods, Geoffrey Corlett  
J.A. KEMP & CO.  
14 South Square  
Gray's Inn  
London WC1R 5LX (GB)

(72) Inventors:  
• Onikubo, Toshikazu  
3-13, Kyobashi 2-chome, Chuo-ku, Tokyo (JP)

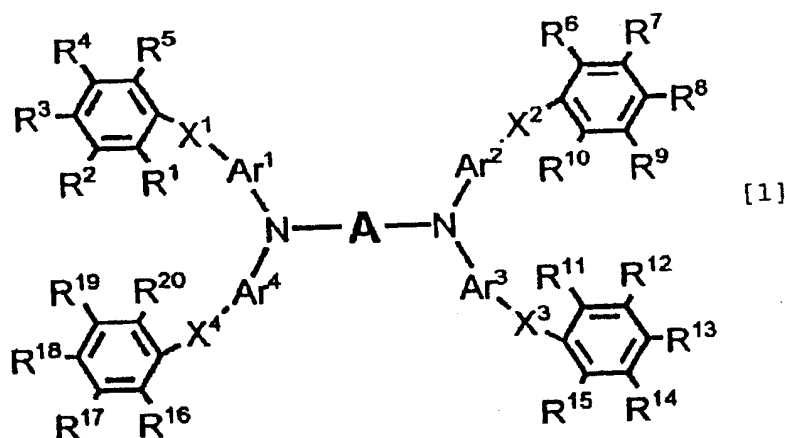
(54) **Light-emitting material for organo-electroluminescence device and organic electroluminescence device**

(57) A light-emitting material which serves to emit light having a high brightness and is almost free of deterioration in light emission, and an organic EL device for which the light-emitting material is adapted, the material having the formula (1)

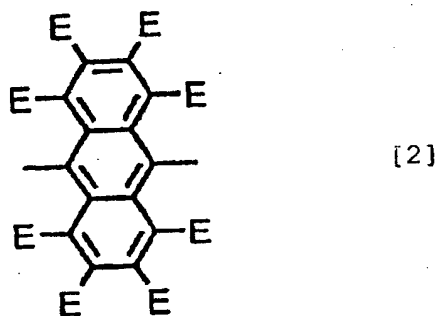


EP 0 866 110 A1

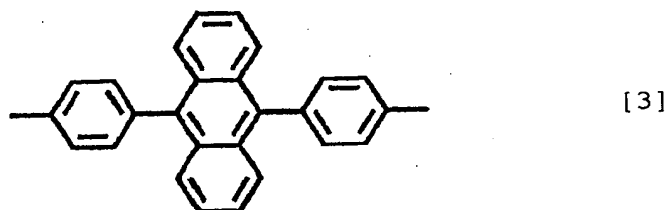
**BEST AVAILABLE COPY**



wherein A is a substituted or non-substituted aromatic group, a substituted or non-substituted fused aromatic group (excluding a group of the formula [2] shown below), a substituted or non-substituted hetero-aromatic group, a substituted or non-substituted fused hetero-aromatic group or a divalent group (excluding a group of the formula [3] shown below) in which 2 to 10 identical or different groups out of the above groups are bonded to each other directly or through at least one of an oxygen atom, a nitrogen atom, a sulfur atom, a linear structural unit having 1 to 20 carbon atoms and optionally containing a hetero-atom, or a non-aromatic ring structural unit, each of Ar<sup>1</sup> to Ar<sup>4</sup> is independently a substituted or non-substituted aromatic group or a substituted or non-substituted fused aromatic group, each of X<sup>1</sup> to X<sup>4</sup> is independently -O-, -S-, >C=O, >SO<sub>2</sub>, -(C<sub>x</sub>H<sub>2x</sub>)-O-(C<sub>y</sub>H<sub>2y</sub>)- (in which each of x and y is an integer of 0 to 20, while x + y = 0 in no case), a substituted or non-substituted alkylidene group having 2 to 20 carbon atoms, a substituted or non-substituted alkylene group having 2 to 20 carbon atoms or a substituted or non-substituted divalent alicyclic residue, and each of R<sup>1</sup> to R<sup>20</sup> is independently a hydrogen atom, a halogen atom, a substituted or non-substituted alkyl group, a substituted or non-substituted alkoxy group, a substituted or non-substituted aromatic group, a substituted or non-substituted hetero-aromatic group or a substituted or non-substituted amino group (provided that adjacent groups of R<sup>1</sup> to R<sup>5</sup>, R<sup>6</sup> to R<sup>10</sup>, R<sup>11</sup> to R<sup>15</sup>, or R<sup>16</sup> to R<sup>20</sup> may bond to each other to form a fresh ring).



wherein E is a hydrogen atom or any adjacent two E's may bond to each other to form a fresh six-membered ring.



Further, according to the present invention, there is provided a light-emitting material of the formula [4] for an

## Detailed Description of the Invention

In the compounds of the formulae [1], [4] and [5] in the present invention, A is a substituted or non-substituted divalent aromatic group, a substituted or non-substituted divalent fused aromatic group, a substituted or non-substituted divalent hetero-aromatic group, a substituted or non-substituted divalent fused hetero-aromatic group or a divalent group in which 2 to 10 identical or different groups out of the above groups are bonded to each other directly or through an oxygen atom, a nitrogen atom, a sulfur atom, or a linear or non-aromatic structural unit containing a hetero-atom. Those sites of A which bond to nitrogen atoms have a ring structure each.

Specific examples of A include substituted or non-substituted divalent aromatic or fused aromatic group residues of benzene, toluene, xylene, ethylbenzene, naphthalene, anthracene (excluding a case where 9- and 10-positions thereof bond), phenanthrene, fluorene, pyrene, chrysene, naphthacene, perylene, azulene, fluorenone, anthraquinone, dibenzosuberone and tetracyanoquinodimethane, and divalent hetero-aromatic or fused-hetero-aromatic residues of furan, thiophene, pyrrole, pyridine, pyrone, oxazole, pyrazine, oxadiazole, triazole, thiadiazole, indole, quinoline, isoquinoline, carbazole, acridine, thioxanthone, coumarin, acridone, diphenylenesulfone, quinoxaline, benzothiazole, phenazine, phenanthroline, phenothiazine, quinacridone, flavanthrone and indanthrone. Further, A includes divalent residues having a structure in which at least 2 identical or different ring-structural units bond, such as divalent residues of biphenyl, terphenyl, binaphthyl, bifuorenylidene, bipyridine, biquinoline, flavone, phenyltriazine, bisbenzothiazole, bithiophene, phenylbenzotriazole, phenylbenzimidazole, phenylacridine, bis(benzooxazolyl)thiophene, bis(phenyloxazolyl)benzene, biphenylphenyloxadiazole, diphenylbenzoquinone, diphenylisobenzofuran, diphenylpyridine, stilbene, dibenzyl, diphenylmethane, bis(phenylisopropyl)benzene, diphenylfluorene, diphenylhexafluoropropane, dibenzyl naphthyl ketone, dibenzylidenecyclohexanone, distylynaphthalene, (phenylethyl)benzyl naphthalene, diphenyl ether, methyldiphenylamine, benzophenone, phenylbenzoate, diphenyl urea, diphenyl sulfide, diphenyl sulfone, diphenoxybiphenyl, bis(phenoxyphenyl) sulfone, bis(phenoxyphenyl)propane, diphenoxybenzene, ethylene glycol diphenyl ether, neopentyl glycol diphenyl ether, dipicolylamine and dipyridylamine.

Table 1 shows specific examples of chemical structure of A in the light-emitting material of the present invention, although A shall not be limited thereto.

Table 1 (continued)

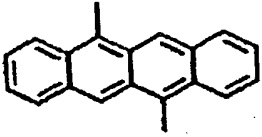
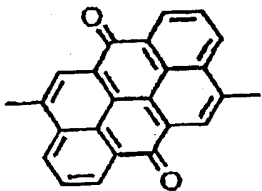
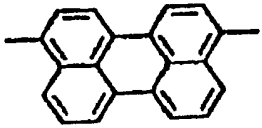
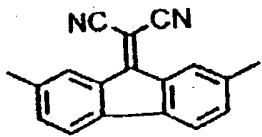
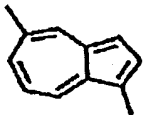
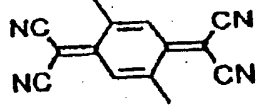
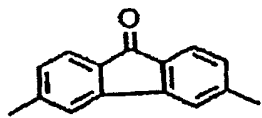
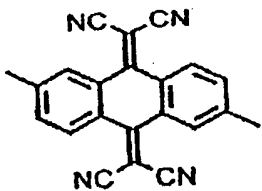
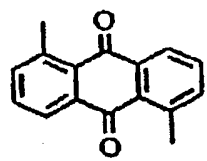

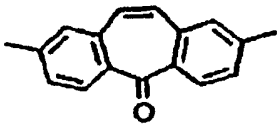

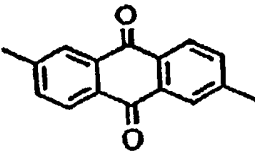
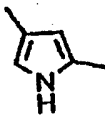
Divalent group	Chemical structure (-A-)	Divalent group	Chemical structure (-A-)
(A-16)		(A-23)	
(A-17)		(A-24)	
(A-18)		(A-25)	
(A-19)		(A-26)	
(A-20)		(A-27)	
(A-21)		(A-28)	
(A-22)		(A-29)	

Table 1 (continued)

Divalent group	Chemical structure (-A-)	Divalent group	Chemical structure (-A-)
(A-46)		(A-54)	
(A-47)		(A-55)	
(A-48)		(A-56)	
(A-49)		(A-57)	
(A-50)		(A-58)	
(A-51)		(A-59)	
(A-52)		(A-60)	
(A-53)			

Table 1 (continued)

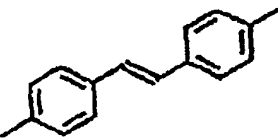
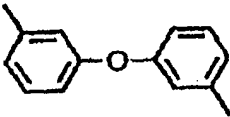
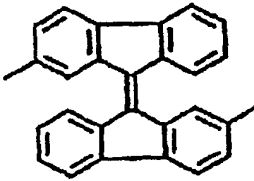
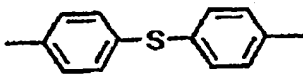
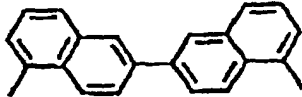
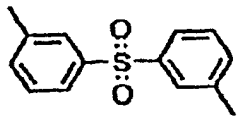
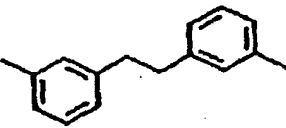
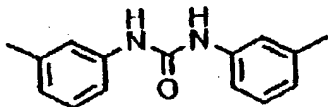
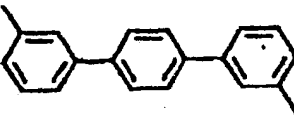
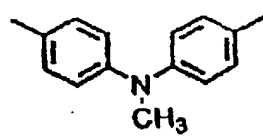
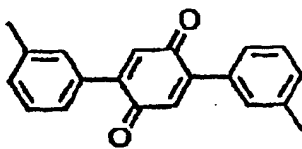
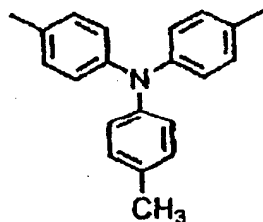
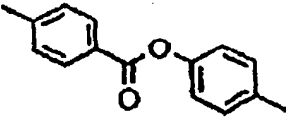
Divalent group	Chemical structure (-A-)	Divalent group	Chemical structure (-A-)
(A-75)		(A-82)	
(A-76)		(A-83)	
(A-77)		(A-84)	
(A-78)		(A-85)	
(A-79)		(A-86)	
(A-80)		(A-87)	
(A-81)			

Table 1 (continued)

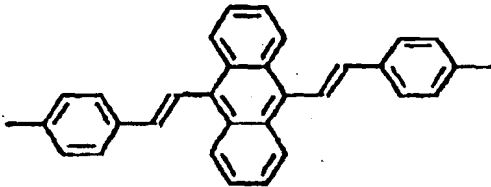
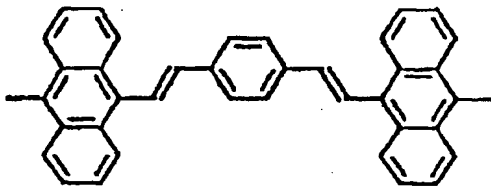
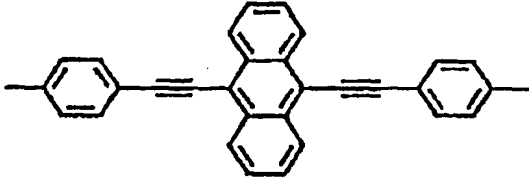
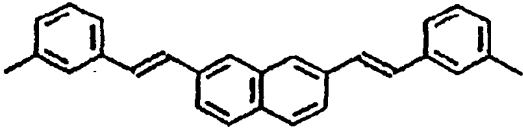
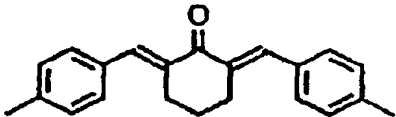
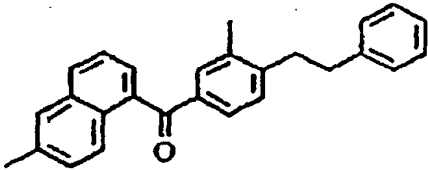
Divalent group	Chemical structure (-A-)
(A-103)	
(A-104)	
(A-105)	
(A-106)	
(A-107)	
(A-108)	

Table 1 (continued)

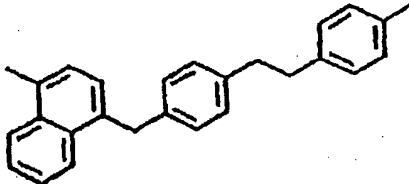
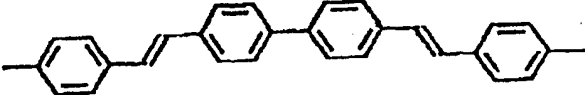
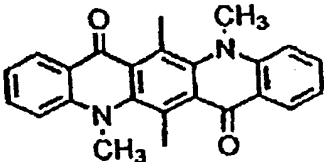
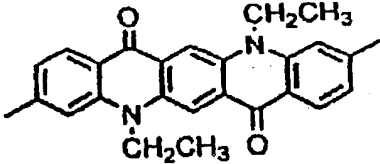
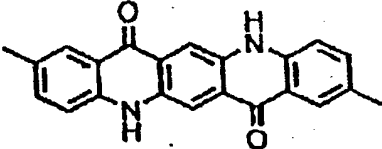
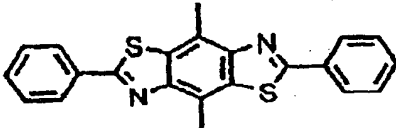
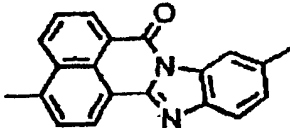
Divalent group	Chemical structure (-A-)
(A-117)	
(A-118)	
(A-119)	
(A-120)	
(A-121)	
(A-122)	
(A-123)	



Table 1 (continued)

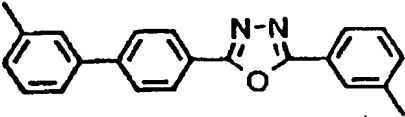
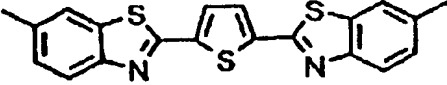

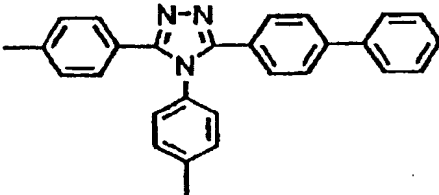
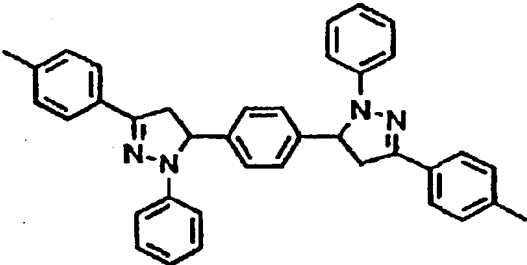
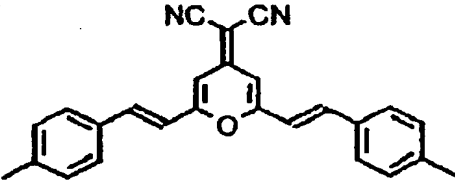
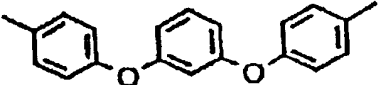
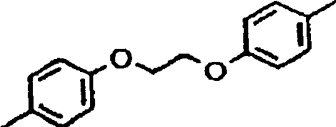
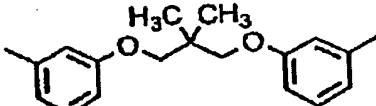
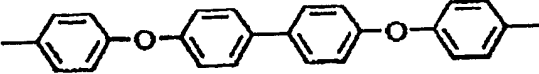
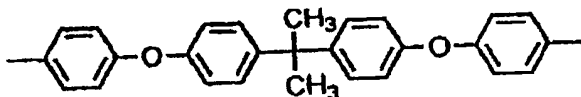
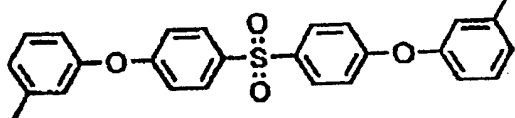
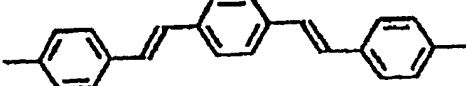
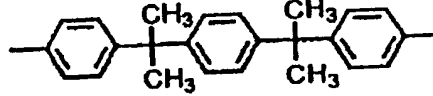
Divalent group	Chemical structure (-A-)
(A-130)	
(A-131)	
(A-132)	
(A-133)	
(A-134)	
(A-135)	

Table 1 (continued)

Divalent group	Chemical structure (-A-)
(A-109)	
(A-110)	
(A-111)	
(A-112)	
(A-113)	
(A-114)	
(A-115)	
(A-116)	

In the compound of the formula [1] in the present invention, each of Ar<sup>1</sup> to Ar<sup>4</sup> is independently a substituted or

Table 2

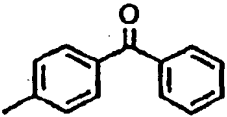
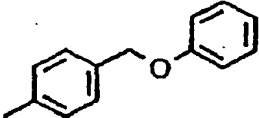
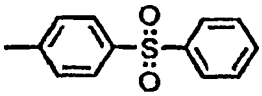
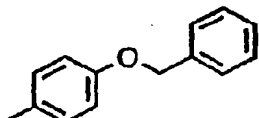
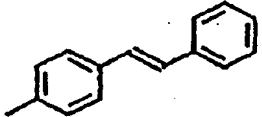
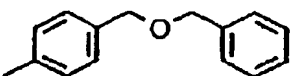
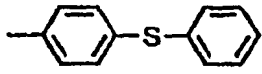
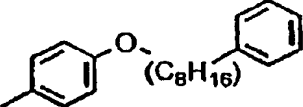
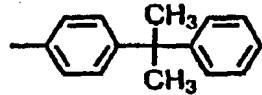
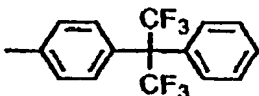
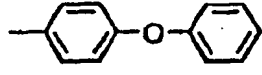
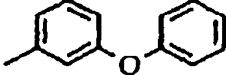
Mono-valent group	Chemical structure	Mono-valent group	Chemical structure
(B-1)		(B-7)	
(B-2)		(B-8)	
(B-3)		(B-9)	
(B-4)		(B-10)	
(B-5)		(B-11)	
(B-6)		(B-12)	

Table 2 (continued)

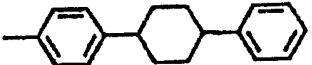
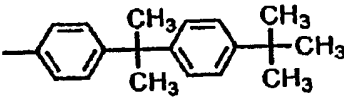
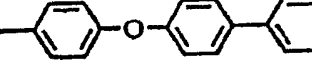
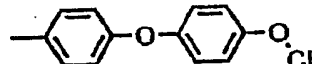
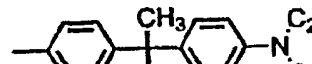
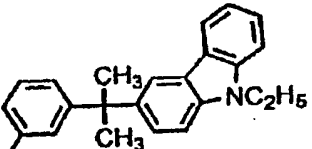
Mono-valent group	Chemical structure
(B-25)	
(B-26)	
(B-27)	
(B-28)	
(B-29)	
(B-30)	

Table 3

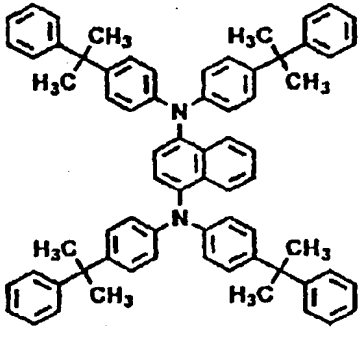
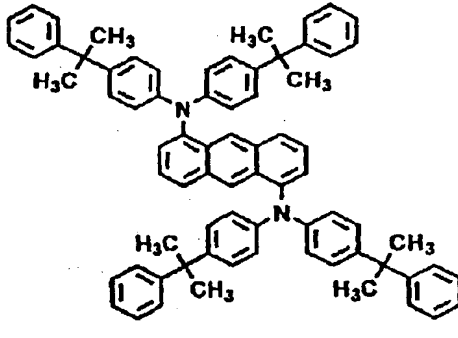
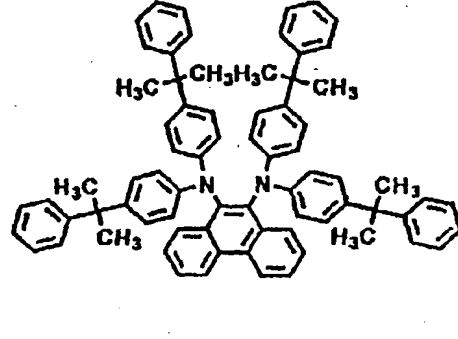
compound	Chemical structure
化合物	化学構造
(1)	
(2)	
(3)	

Table 3 (continued)

compound	Chemical structure
(7)	
(8)	
(9)	

Table 3 (continued)

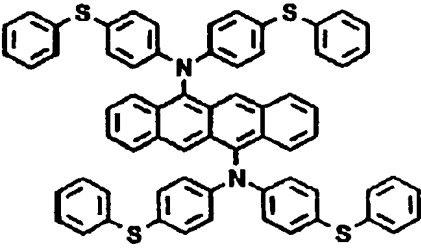
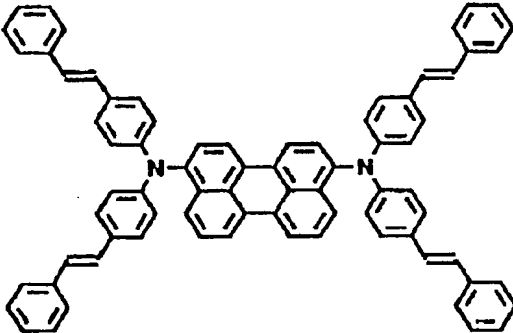
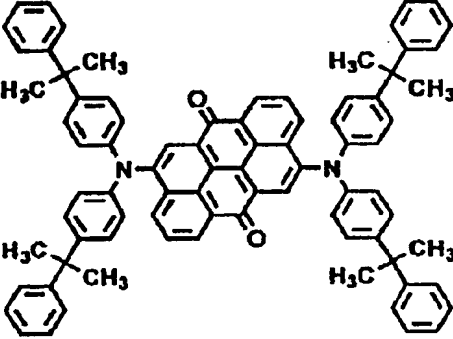
compound	Chemical structure
(13)	 <chem>c1ccc(cc1)S(c2ccc(cc2)N(c3ccc(cc3)S(c4ccccc4)cc4)cc3)c5ccc6cc7ccccc7cc6cc5</chem>
(14)	 <chem>c1ccc(cc1)/C=C/c2ccc(cc2)N(c3ccc4cc5ccccc5cc4cc3)c6ccc7cc8ccccc8cc7cc6</chem>
(15)	 <chem>CC(C)(c1ccccc1)c2ccc(cc2)N(c3ccc4c5c6c7c(=O)c8ccccc8c4c5cc6N(c9ccc10c(c3)C(C)(C)c11ccccc11)cc9)cc4</chem>

Table 3 (continued)

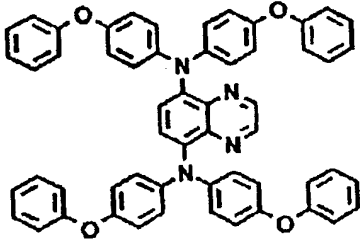
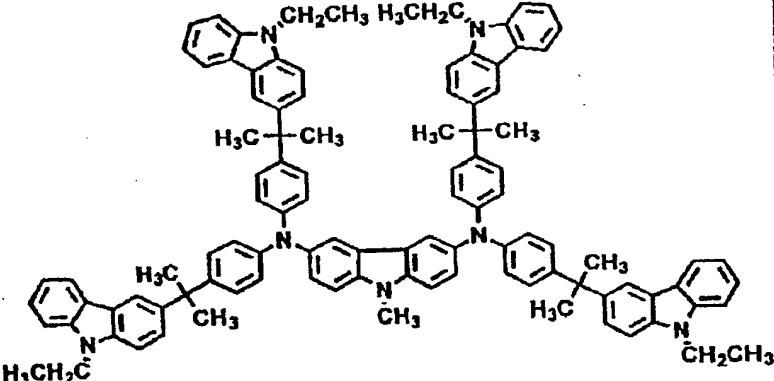
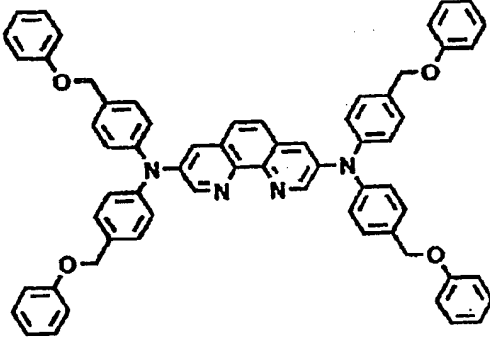
compound	Chemical structure
(19)	
(20)	
(21)	



Table 3 (continued)

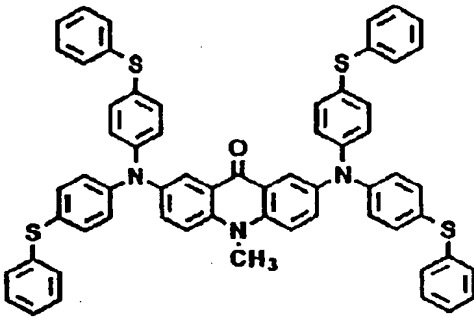
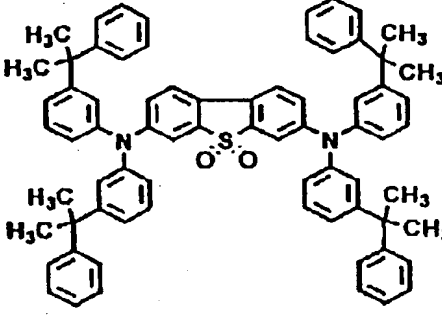
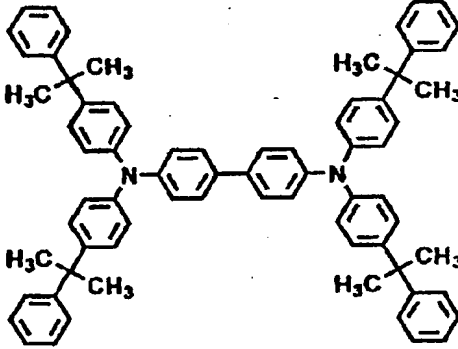
compound	Chemical structure
(25)	
(26)	
(27)	

Table 3 (continued)

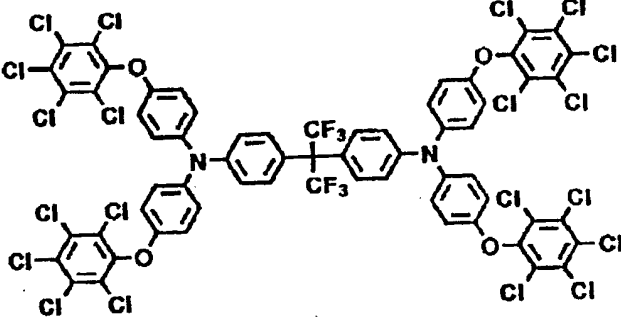
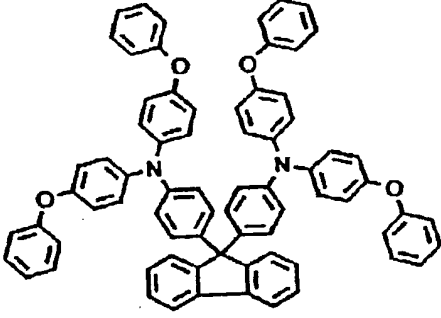
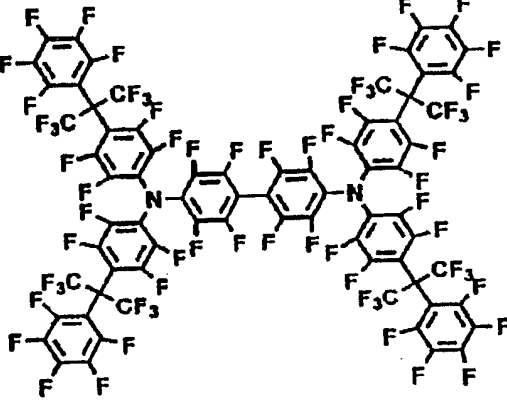
compound	Chemical structure
(31)	
(32)	
(33)	

Table 3 (continued)

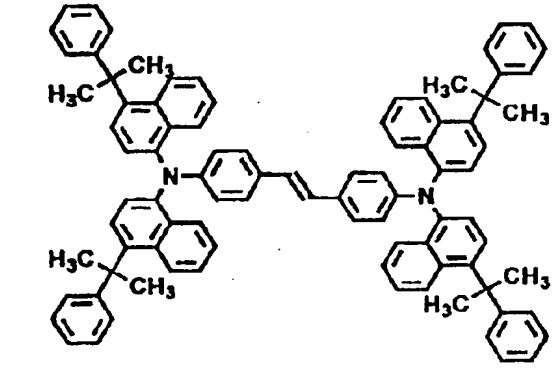
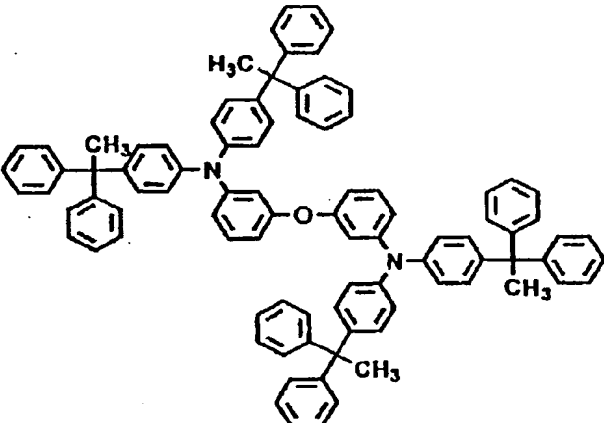
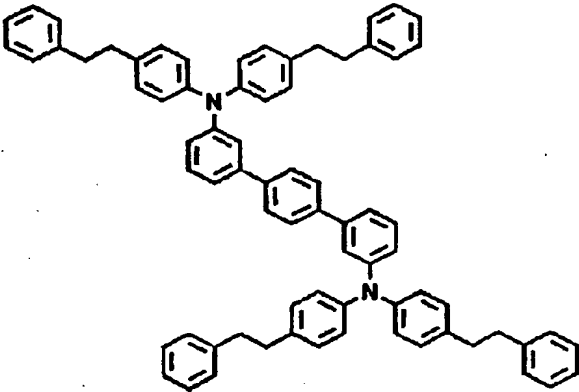
compound	Chemical structure
(37)	
(38)	
(39)	

Table 3 (continued)

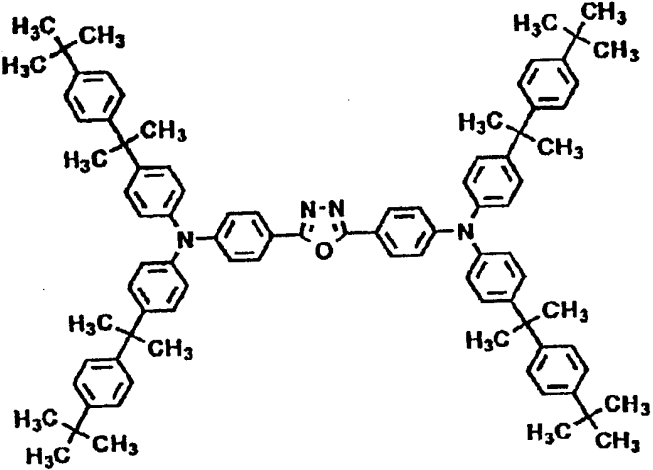
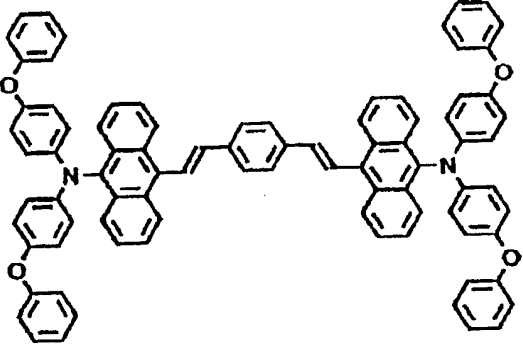
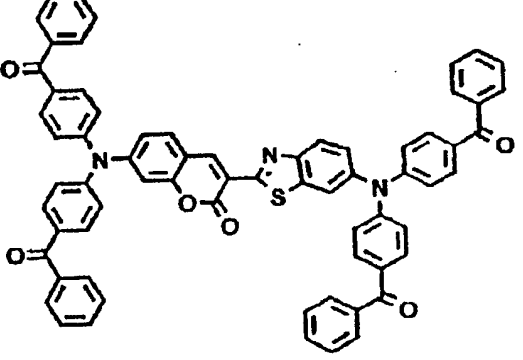
compound	Chemical structure
(43)	
(44)	
(45)	

Table 3 (continued)

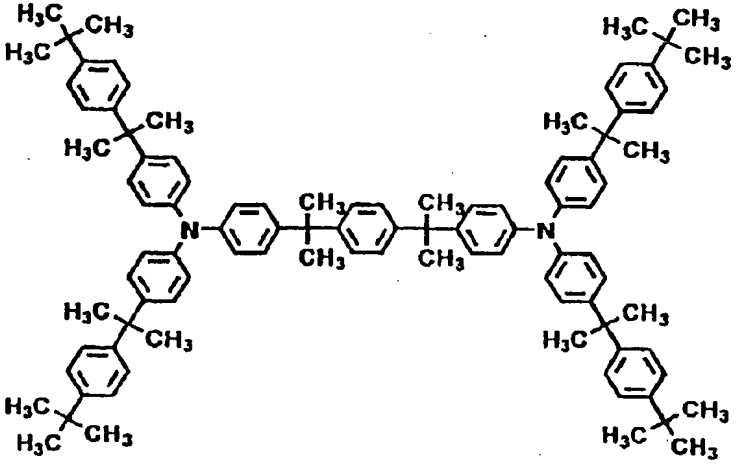
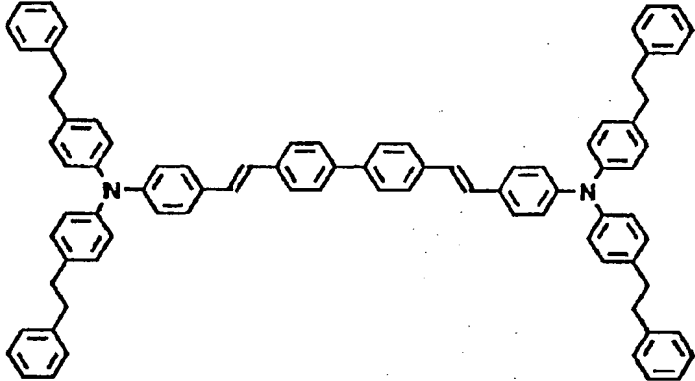
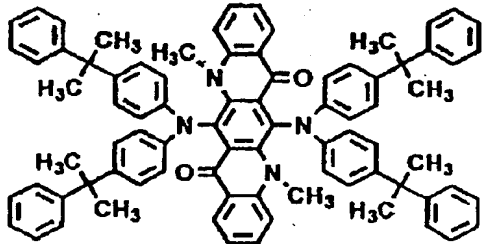
compound	Chemical structure
(49)	
(50)	
(51)	

Table 3 (continued)

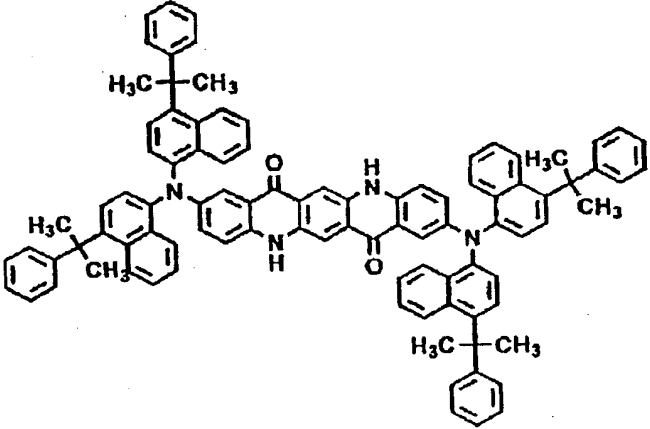
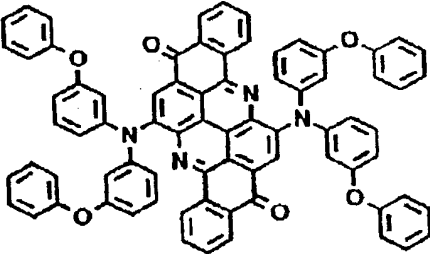
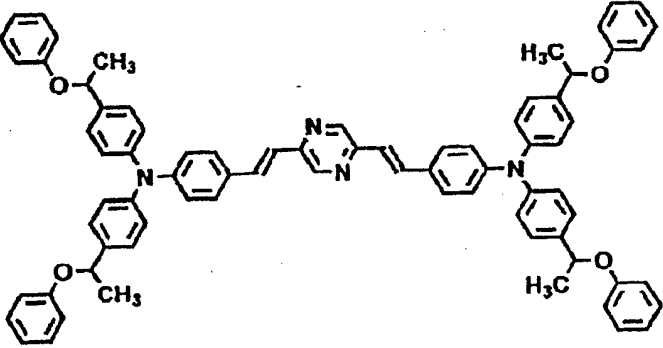
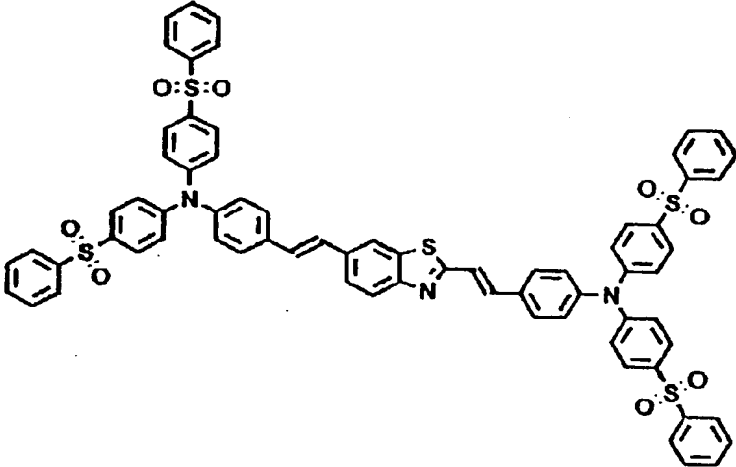
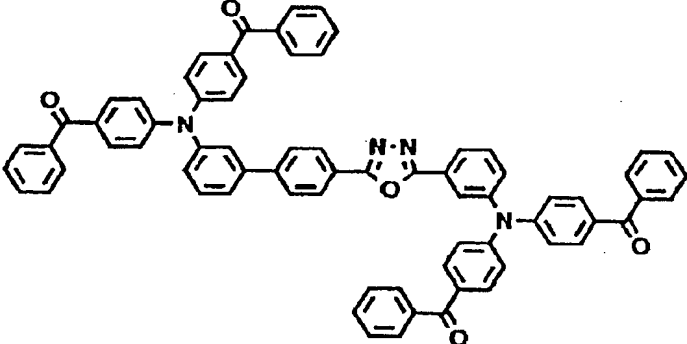
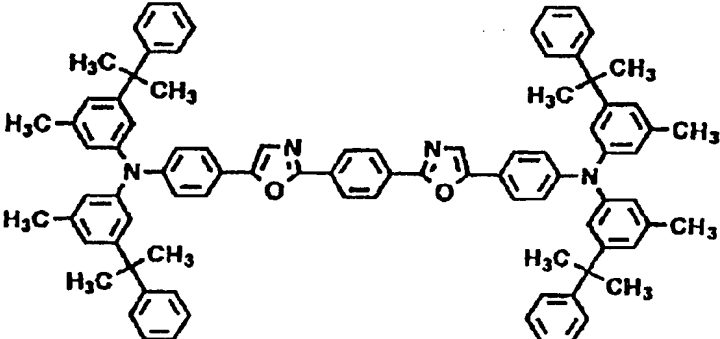
compound	Chemical structure
(55)	
(56)	
(57)	

Table 3 (continued)

compound	Chemical structure
(61)	
(62)	
(63)	

5

55



Table 3 (continued)

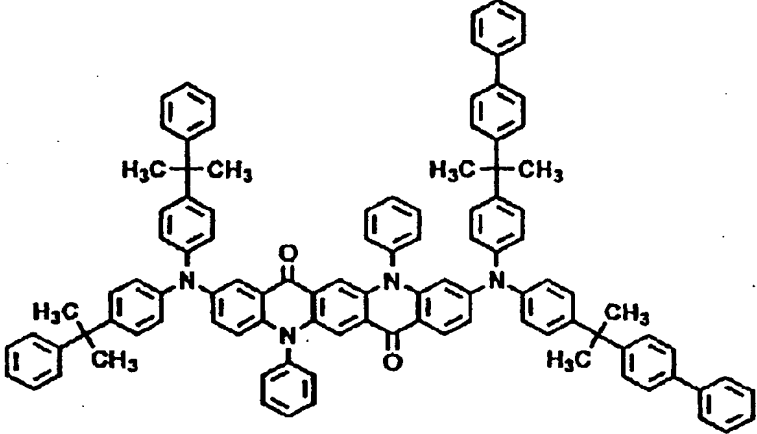
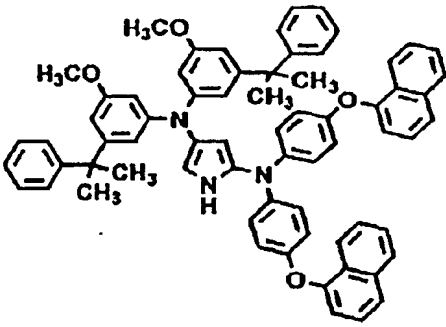
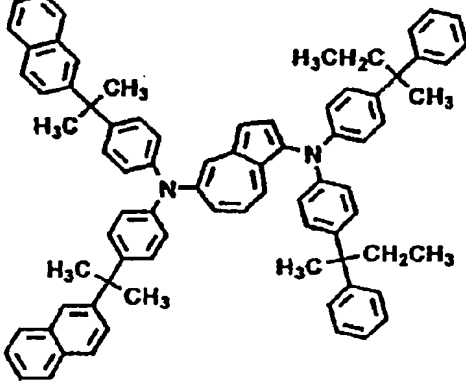
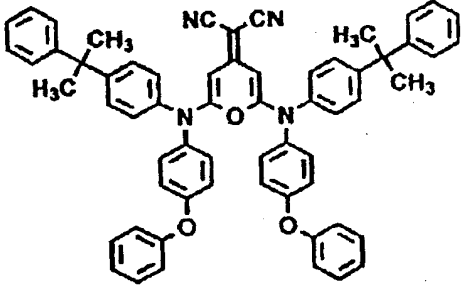
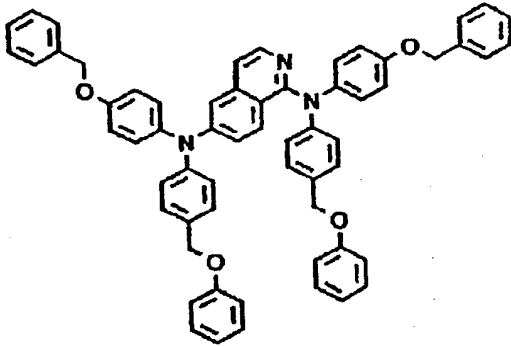
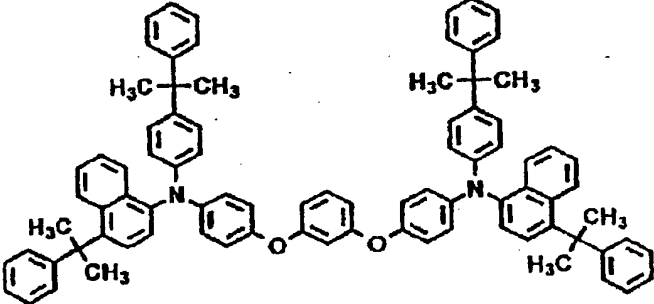
compound	Chemical structure
(73)	
(74)	
(75)	

Table 3 (continued)

compound	Chemical structure
(79)	
(80)	
(81)	

The light-emitting material of the present invention (compounds of the present invention) is a compound having intense fluorescence in a solid state, and is excellent in electric-field-applied light emission characteristic. Further, the light-emitting material of the present invention is excellent in the characteristic of injection of holes from a metal electrode and the property of transportation of holes, and it is also excellent in the characteristics of injection of electrons from a metal electrode and the property of transportation of electrons. It can be therefore effectively used as a light-emitting material, and further, it can be used in combination with other hole-transporting material, an electron-transporting material or a dopant without any problem.

An organic EL device is a device having a structure in which a mono- or multi-layered organic thin layer is formed between an anode and a cathode. In a mono-layered device, a light-emitting layer is formed between the anode and the cathode. The light-emitting layer contains a light-emitting material, and in addition thereto, it may contain a hole-injecting material for transporting holes injected from the anode to the light-emitting material, or an electron-injecting material for transporting electrons injected from the cathode to the light-emitting material. The light-emitting material of the present invention has a remarkably high light emission quantum efficiency, high capability of transporting holes and high capability of transporting electrons and can form a uniform thin film. The light-emitting layer can be therefore formed of the light-emitting material of the present invention alone. A multi-layered organic EL device has one of laminated-layer structures, for example, of (anode/hole-injecting zone/light-emitting layer/cathode), (anode/light-emitting layer/electron-injecting layer/zone) and (anode/hole-injecting zone/light-emitting layer/electron-injecting zone/cathode). The light-emitting material of the present invention (compounds of the present invention) can be used in a light-emitting layer due to its characteristic of high light emission, property of injecting holes, capability of transporting holes and property of injecting electrons and capability of transporting electrons.

In addition to the material of the present invention, the light-emitting layer may contain a known light-emitting material, a known dopant, a known hole-injecting material or a known electron-injecting material as required. In the organic EL device, a decrease in the brightness and life caused by quenching can be prevented by forming it as a multi-layered structure. The light-emitting material, a dopant, a hole-injecting material and an electron-injecting material may be used in combination as required. Further, a dopant can improve the light emission brightness and the light emission efficiency, and can attain the red or blue light emission. Further, each of the hole-injecting zone, the light-emitting layer and the electron-injecting zone may have the layer structure of at least two layers. In the hole-injecting zone in this case, a layer to which holes are injected from an electrode is called "hole-injecting layer", and a layer which receives holes from the hole-injecting layer and transport the holes to a light-emitting layer is called "hole-transporting layer". In the electron-injecting zone, a layer to which electrons are injected from an electrode is called "electron-injecting layer", and a layer which receives electrons from the electron-injecting layer and transports the electrons to a light-emitting layer is called "electron-transporting layer". These layers are selected and used depending upon factors such as the energy level and heat resistance of materials and adhesion to an organic layer or metal electrode.

The light-emitting material or the dopant which may be used in the light-emitting layer together with the light-emitting material of the present invention includes anthracene, naphthalene, phenanthrene, pyrene, tetracene, coronene, chrysene, fluorescein, perylene, phthaloperylene, naphthaloperylene, perinone, phthaoperinone, naphthaloperinone, diphenylbutadiene, tetraphenylbutadiene, coumarine, oxadiazole, aldazine, bisbenzoxazoline, bisstyryl, pyrazine, cyclopentadiene, quinoline metal complex, aminoquinoline metal complex, benzoquinoline metal complex, imine, diphenylethylene, vinyl anthracene, diaminocarbazole, pyran, thiopyran, polymethine, merocyanine, an imidazole-chelated oxynoid compound, quinacridone, rubrene, and fluorescent dyestuffs for a dyestuff laser or for brightening, although the above material shall not be limited to these.

The light-emitting material of the present invention and the above compound that can be used in a light-emitting layer may be used in any mixing ratio for forming a light-emitting layer. That is, the light-emitting material of the present invention may be a main component for forming a light-emitting layer, and it may be a doping material in other main material, depending upon a combination of the above compound with the light-emitting material of the present invention.

The hole-injecting material is selected from compounds which are capable of transporting holes, are capable of receiving holes from the anode, have an excellent effect of injecting holes to a light-emitting layer or a light-emitting material, prevent the movement of excitons generated in a light-emitting layer to an electron-injecting zone or an electron-injecting material and have the excellent capability of forming a thin film. Specific examples of the above hole-injecting material include a phthalocyanine derivative, a naphthalocyanine derivative, a porphyrin derivative, oxazole, oxadiazole, triazole, imidazole, imidazolone, imidazolthione, pyrazoline, pyrazolone, tetrahydroimidazole, oxazole, oxadiazole, hydrazone, acylhydrazone, polyaryalkane, stilbene, butadiene, benzidine type triphenylamine, styrylamine type triphenylamine, diamine type triphenylamine, derivatives of these, and polymer materials such as polyvinylcarbazole, polysilane and an electroconducting polymer. However, the hole-transporting material shall not be limited to the above materials.

In the organic EL device of the present invention, the hole-injecting material which is more effective is an aromatic tertiary amine derivative or a phthalocyanine derivative. Although not specially limited, specific examples of the tertiary amine derivative include triphenylamine, tritolylamine, tolyldiphenylamine, N,N'-diphenyl-N,N'-(3-methylphenyl)-1,1-bi-

large, inefficiently, a high voltage is required to achieve predetermined emission of light. When the layer thickness is too small, the layer is liable to have a pinhole, etc., so that sufficient light emission brightness is hard to obtain when an electric field is applied. Generally, the thickness of each layer is preferably in the range of from 5 nm to 10  $\mu\text{m}$ , more preferably 10 nm to 0.2  $\mu\text{m}$ .

In the wet film forming method, a material for forming an intended layer is dissolved or dispersed in a proper solvent such as ethanol, chloroform, tetrahydrofuran and dioxane, and a thin film is formed from the solution or dispersion. The solvent shall not be limited to the above solvents. For improving the film formability and preventing the occurrence of pinholes in any layer, the above solution or dispersion for forming the layer may contain a proper resin and a proper additive. The resin that can be used includes insulating resins such as polystyrene, polycarbonate, polyarylate, polyester, polyamide, polyurethane, polysulfone, polymethyl methacrylate, polymethyl acrylate and cellulose, copolymers of these, photoconductive resins such as poly-N-vinylcarbazole and polysilane, and electroconducting polymers such as polythiophene and polypyrrole. The above additive includes an antioxidant, an ultraviolet absorbent and a plasticizer.

When the light-emitting material of the present invention is used in a light-emitting layer of an organic EL device, an organic EL device can be improved in organic EL device characteristics such as light emission efficiency and maximum light emission brightness. Further, the organic EL device of the present invention is remarkably stable against heat and electric current and gives a practically usable light emission brightness at a low actuation voltage. The deterioration which is a big problem of conventional devices can be remarkably decreased.

The organic EL device of the present invention has significant industrial values since it can be adapted for a flat panel display of an on-wall television set, a flat light-emitting device, a light source for a copying machine or a printer, a light source for a liquid crystal display or counter, a display signboard and a signal light.

The material of the present invention can be used in the fields of an organic EL device, an electrophotographic photoreceptor, a photoelectric converter, a solar cell, an image sensor, and the like.

#### Examples

The present invention will be explained more in detail with reference to Examples hereinafter.

#### Example 1

Compound (1) in Table 3 as a light-emitting material, 2,5-bis(1-naphthyl)-1,3,4-oxadiazole and a polycarbonate resin (Panlite K-1300, supplied by Teijin Kasei) in a weight ratio of 5:3:2 were dissolved in tetrahydrofuran, and the solution was spin-coated on a cleaned glass substrate with an ITO electrode to form a light-emitting layer having a thickness of 100 nm. An electrode having a thickness of 150 nm was formed thereon from a magnesium/indium alloy having a magnesium/indium mixing ratio of 10/1, to obtain an organic EL device. The device had the following light emission characteristics. The device showed blue light emission having a brightness of 90 ( $\text{cd}/\text{m}^2$ ) at a direct current voltage of 5 V, a maximum brightness of 1,500 ( $\text{cd}/\text{m}^2$ ) and a light emission efficiency of 0.50  $\text{lm}/\text{W}$ .

#### Example 2

Compound (2) in Table 3 was vacuum-deposited on a cleaned glass substrate with an ITO electrode to form a light-emitting layer having a thickness of 100 nm. An electrode having a thickness of 100 nm was formed thereon from a magnesium/silver alloy having a magnesium/silver mixing ratio of 10/1, to obtain an organic EL device. The light-emitting layer was formed by deposition under a vacuum of  $10^{-6}$  Torr at a substrate temperature of room temperature. The device showed green light emission having a brightness of 260 ( $\text{cd}/\text{m}^2$ ) at a direct current voltage of 5 V, a maximum brightness of 800 ( $\text{cd}/\text{m}^2$ ) and a light emission efficiency of 0.60  $\text{lm}/\text{W}$ .

#### Example 3

Compound (3) in Table 3 was dissolved in methylene chloride tetrahydrofuran, and the solution was spin-coated on a cleaned glass substrate with an ITO electrode to form a light-emitting layer having a thickness of 50 nm. Then, aluminum bis(2-methyl-8-quinolate)(2-naphtholate) was vacuum-deposited to form an electron-injecting layer having a thickness of 10 nm, and an electrode having a thickness of 100 nm was formed thereon from a magnesium/aluminum alloy having a magnesium/aluminum mixing ratio of 10/1, to obtain an organic EL device. The light-emitting layer and the electron-injecting layer were formed by deposition under a vacuum of  $10^{-6}$  Torr at a substrate temperature of room temperature. The device showed bluish green light emission having a brightness of 200 ( $\text{cd}/\text{m}^2$ ) at a direct current voltage of 5 V, a maximum brightness of 12,000 ( $\text{cd}/\text{m}^2$ ) and a light emission efficiency of 1.2  $\text{lm}/\text{W}$ .

Table 4

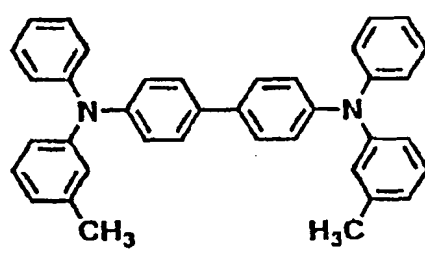
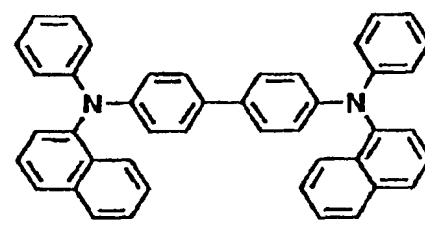
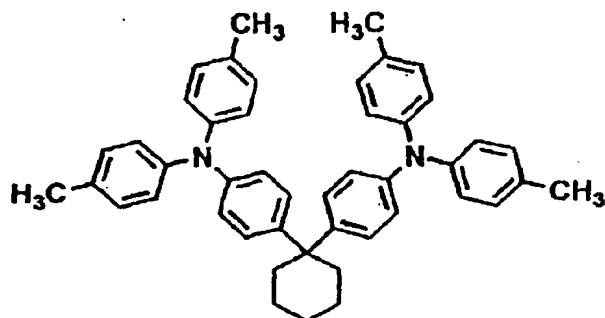
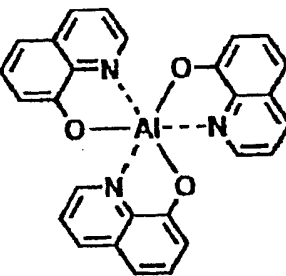
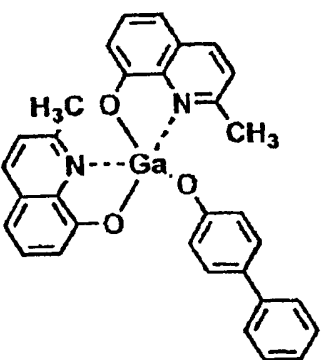
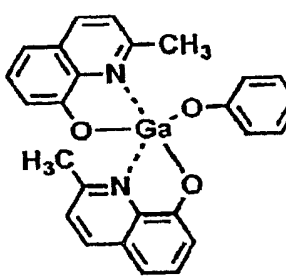
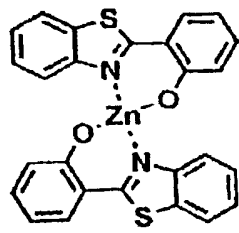
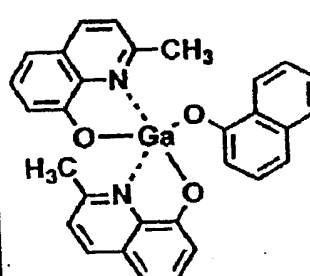
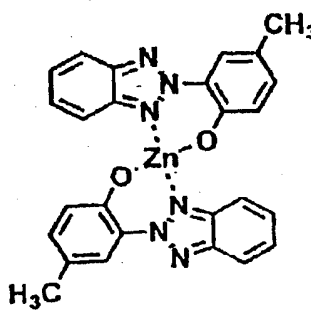
compound	Chemical structure
(H-1)	 <chem>Cc1ccc(N(c2ccccc2)c3ccc(cc3)-c4ccc(cc4)N(c5ccccc5)c6ccc(C)cc6)cc1</chem>
(H-2)	 <chem>c1ccc2c(c1)c3ccccc3N(c4ccccc4)c5ccc(cc25)-c6ccc(cc6)N(c7ccccc7)c8cccc9ccccc89</chem>
(H-3)	 <chem>Cc1ccc(N(c2ccc(C)cc2)c3ccccc3C4CCCCC4c5ccccc5N(c6ccc(C)cc6)c7ccc(C)cc7)cc1</chem>

Table 4 (continued)

compound	Chemical structure	Compound	Chemical structure
(E-1)		(E-4)	
(E-2)		(E-5)	
(E-3)		(E-6)	

EP 0 866 110 A1

Table 5 (continued)

	Ex.	Hole-injecting material (Table 4)	Light-emitting material (Table 3)	Electron-injecting material (Table 4)	Light emission brightness (cd/m <sup>2</sup> )	Maximum light emission brightness (cd/m <sup>2</sup> )	Maximum light emission efficiency (1m/W)
5	40	(H-1)	(41)	(E-2)	320	19,600	2.6
	41	(H-6)	(42)	(E-4)	770	45,200	4.7
10	42	(H-5)	(43)	(E-5)	400	35,100	2.9
	43	(H-2)	(44)	(E-1)	730	26,100	2.2
	44	(H-1)	(45)	(E-4)	550	41,800	4.8
	45	(H-2)	(46)	(E-4)	440	29,900	2.9
15	46	(H-3)	(47)	(E-4)	290	9,700	1.6
	47	(H-6)	(48)	(E-4)	270	8,100	2.4
	48	(H-5)	(49)	(E-6)	690	15,000	1.0
20	49	(H-2)	(50)	(E-5)	330	48,600	5.1
	50	(H-3)	(51)	(E-6)	490	57,300	6.1
	51	(H-6)	(52)	(E-5)	280	52,800	5.6
	52	(H-2)	(53)	(E-2)	290	41,000	5.4
25	53	(H-5)	(54)	(E-3)	790	30,900	2.5
	54	(H-3)	(55)	(E-4)	440	47,800	3.9
	55	(H-3)	(56)	(E-6)	280	47,600	4.6
30	56	(H-4)	(57)	(E-2)	760	15,600	1.4
	57	(H-1)	(58)	(E-2)	700	19,000	1.7
	58	(H-4)	(59)	(E-6)	660	14,600	1.8
35	59	(H-3)	(60)	(E-4)	420	31,700	3.7
	60	(H-3)	(61)	(E-5)	560	48,900	4.3
	61	(H-2)	(62)	(E-6)	600	44,800	5.2
	62	(H-4)	(63)	(E-4)	270	39,200	3.7
40	63	(H-5)	(64)	(E-2)	330	20,700	1.6
	64	(H-6)	(65)	(E-1)	690	7,600	1.9
	65	(H-6)	(66)	(E-3)	390	9,800	1.6
45	66	(H-5)	(67)	(E-1)	770	9,200	1.7
	67	(H-4)	(68)	(E-5)	660	9,200	1.3
	68	(H-2)	(69)	(E-6)	700	21,000	2.7
	69	(H-4)	(70)	(E-4)	210	34,100	4.6
50	70	(H-5)	(71)	(E-3)	630	47,300	4.4
	71	(H-6)	(72)	(E-3)	660	29,300	3.5
	72	(H-5)	(73)	(E-4)	510	41,400	5.1
55	73	(H-1)	(74)	(E-2)	690	18,400	1.4
	74	(H-5)	(75)	(E-3)	370	8,200	1.9
	75	(H-4)	(76)	(E-2)	570	22,500	1.6

Examples 87 - 90

An organic EL device was prepared in the same manner as in Example 5 except that the light-emitting layer was replaced with a 30 nm thick light-emitting layer formed by vacuum-depositing Compound (5) in Table 3 and one of Compounds (D-1) to (D-7) in Table 6 in a weight ratio of 100:1. Table 7 shows light emission characteristics of the so-obtained devices. All the organic EL devices obtained in these Examples showed high brightness characteristics, or a maximum brightness of at least 20,000 (cd/m<sup>2</sup>), and gave intended light emission colors.

10

15

20

25

30

35

40

45

50

55



## Examples 90 - 94

An organic EL device was prepared in the same manner as in Example 5 except that the light-emitting layer was replaced with a 30 nm thick light-emitting layer formed by vacuum-depositing Compound (27) in Table 3 and one of Compounds (D-1) to (D-7) in Table 6 in a weight ratio of 100:1. Table 7 shows light emission characteristics of the so-obtained devices. All the organic EL devices obtained in these Examples showed high brightness characteristics, or a maximum brightness of at least 20,000 (cd/m<sup>2</sup>), and gave intended light emission colors.

## Example 95

A hole-injecting material (H-2) was vacuum-deposited on a cleaned glass substrate with an ITO electrode to form a hole-injecting layer having a thickness of 30 nm. Then, 4,4'-bis( $\beta$ , $\beta$ -diphenylvinyl)biphenyl and a light-emitting material (1) in Table 3 for a light-emitting layer were vacuum-deposited in a weight ratio of 100:5 to form a light-emitting layer having a thickness of 30 nm. Further, an electron-injecting material (E-3) was vacuum-deposited to form an electron-injecting layer having a thickness of 30 nm. Then, an electrode having a thickness of 150 nm was formed thereon from a magnesium/silver alloy having an magnesium/silver mixing ratio of 10/1, to obtain an organic EL device. The device showed blue light emission having a brightness of 480 (cd/m<sup>2</sup>) at a direct current voltage of 5 V, a maximum brightness of 28,000 (cd/m<sup>2</sup>) and a light emission efficiency of 3.1 lm/W.

## Examples 96 - 108

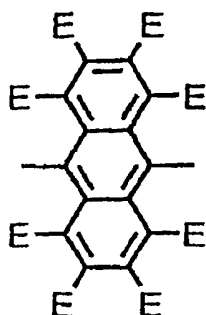
An organic EL device was prepared in the same manner as in Example 95 except that the light-emitting layer was replaced with a 30 nm thick light-emitting layer formed by vacuum-depositing aluminum tris(8-hydroxyquinolate) and one of the light-emitting materials in Table 3 in a weight ratio of 100:3. Table 7 shows light emission characteristics of the so-obtained devices. All the organic EL devices obtained in these Examples showed high brightness characteristics, or a maximum brightness of at least 20,000 (cd/m<sup>2</sup>).

Table 7

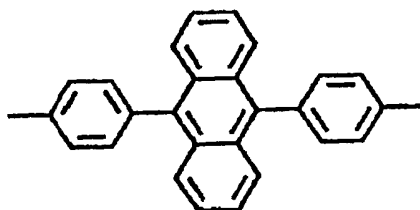
Ex.	Compound (Table 3 or 6)	Light emission brightness (cd/m <sup>2</sup> )	Maximum light emission brightness (cd/m <sup>2</sup> )	Maximum light emission efficiency (lm/W)
87	(D-1)	720	78,400	8.1
88	(D-2)	310	53,700	4.5
89	(D-3)	250	39,800	4.8
90	(D-4)	830	37,100	3.9
91	(D-5)	260	55,200	5.2
92	(D-6)	480	29,200	2.3
93	(D-7)	800	37,800	3.8
94	(D-3)	810	27,700	2.4
96	(2)	390	58,000	6.2
97	(4)	250	29,600	3.4
98	(14)	220	61,800	5.1
99	(15)	160	54,400	3.7
100	(23)	240	46,700	3.8
101	(36)	870	55,200	5.9
102	(41)	560	26,500	4.1
103	(54)	830	35,300	3.9
104	(55)	870	59,200	6.7
105	(58)	210	24,500	2.1
106	(64)	640	23,800	3.1

- A, which can be substituted or non-substituted, is a divalent fused or non fused aromatic group, a hetero-aromatic fused or non fused divalent group, or a divalent group in which from 2 to 10 said divalent aromatic or hetero-aromatic divalent groups, which can be the same or different, are bonded to each other directly or through at least one of an oxygen atom, a nitrogen atom, a sulfur atom, a linear structural unit having 1 to 20 carbon atoms and optionally containing a heteroatom, or a non-aromatic ring structural unit;
- each of Ar<sup>1</sup> to Ar<sup>4</sup> is independently a substituted or non-substituted fused or non fused aromatic group;
- each of X<sup>1</sup> to X<sup>4</sup> is independently -O-, -S-, >C=O, >SO<sub>2</sub>, -(C<sub>x</sub>H<sub>2x</sub>)-O-(C<sub>y</sub>H<sub>2y</sub>)- (in which x and y each independently represent an integer of 0 to 20, while x + y = 0 in no case), a substituted or non-substituted alkylidene group having from 2 to 20 carbon atoms, a substituted or non-substituted alkylene group having 2 to 20 carbon atoms or a substituted or non-substituted divalent alicyclic residue; and
- each of R<sup>1</sup> to R<sup>20</sup> is independently a hydrogen atom, a halogen atom, a substituted or non-substituted alkyl group, a substituted or non-substituted alkoxy group, a substituted or non-substituted aromatic group, a substituted or non-substituted hetero-aromatic group or a substituted or non-substituted amino group, or two adjacent groups of R<sup>1</sup> to R<sup>5</sup>, R<sup>6</sup> to R<sup>10</sup>, R<sup>11</sup> to R<sup>15</sup> and/or R<sup>16</sup> to R<sup>20</sup>, together with the carbon atoms to which they are attached, form a cyclic moiety,

provided that A is not a group of formula [2] or [3]



wherein E is a hydrogen atom or any adjacent two E's may bond to each other to form a six-membered ring,



2. A compound according to claim 1, wherein each of Ar<sup>1</sup> to Ar<sup>4</sup> represent a 1,4 divalent phenyl moiety.
3. A compound of formula [5] suitable for use in an organic electroluminescence device,



European Patent  
Office

## EUROPEAN SEARCH REPORT

Application Number  
EP 98 30 1986

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
X	PATENT ABSTRACTS OF JAPAN vol. 017, no. 704 (C-1146), 22 December 1993 & JP 05 239455 A (RICOH CO LTD), 17 September 1993, * abstract; examples 9,10 * ---	1-10	C09K11/06 H05B33/14 G03G5/06 C07C211/54
X	EP 0 666 298 A (TDK CORP) 9 August 1995 * examples II-8,II-16 * * examples III-15,IV-7 * * examples VI-7 * ---	1,2,4-10	
A	PATENT ABSTRACTS OF JAPAN vol. 096, no. 006, 28 June 1996 & JP 08 053397 A (TOYO INK MFG CO LTD), 27 February 1996, * abstract; example 9 * ---	1-10	
P,A	EP 0 786 926 A (TOYO INK MFG CO) 30 July 1997 * page 2, line 1 - page 3, line 25; examples (23-40); ---	1-10	TECHNICAL FIELDS SEARCHED (Int.Cl.6)
A	STICKLEY K R ET AL: "Cation Radicals of 1,3,5-Tris(diarylamino)benzenes" TETRAHEDRON LETTERS, vol. 36, no. 10, 6 March 1995, page 1585-1588 XP004028538 * example 2D * ---	1-3	C09K H05B C07C G03G
A	PATENT ABSTRACTS OF JAPAN vol. 097, no. 002, 28 February 1997 & JP 08 259937 A (TOYO INK MFG CO LTD), 8 October 1996, * abstract; example 17 * ---	1-10	
-/--			
The present search report has been drawn up for all claims			
Place of search		Date of completion of the search	Examiner
THE HAGUE		24 June 1998	Shade, M
CATEGORY OF CITED DOCUMENTS			
X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document	

EPO FORM 1503 03/82 (P04C01)

**THIS PAGE BLANK (USPTO)**

**This Page is Inserted by IFW Indexing and Scanning  
Operations and is not part of the Official Record**

**BEST AVAILABLE IMAGES**

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

- ☐ **BLACK BORDERS**
- ☐ **IMAGE CUT OFF AT TOP, BOTTOM OR SIDES**
- ☐ **FADED TEXT OR DRAWING**
- ☐ **BLURRED OR ILLEGIBLE TEXT OR DRAWING**
- ☐ **SKEWED/SLANTED IMAGES**
- ☐ **COLOR OR BLACK AND WHITE PHOTOGRAPHS**
- ☐ **GRAY SCALE DOCUMENTS**
- ☐ **LINES OR MARKS ON ORIGINAL DOCUMENT**
- ☐ **REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY**
- ☐ **OTHER:** \_\_\_\_\_

**IMAGES ARE BEST AVAILABLE COPY.**

**As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.**

**THIS PAGE BLANK (USPTO)**